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An application of the atomic distribution function of liquid mercury

Robert Lee Choate

Department:

AN APPLICATION OF THE ATOMIC DISTRIBUTION FUNCTION OF LIQUID MERCURY

BY

ROBERT L. CHOATE

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R. L. C.

CONTENTS

LIST OF ILLUSTRATIONS

LIST OF TABLES

INTRODUCTION

The study of liquids and the knowledge of an adequate description of liquid structure have not advanced as rapidly as it has in the case of solids and gases. In a perfect crystal, the atoms or molecules are held in equilibrium positions by strong intermolecular forces. There are vibrations about these positions, but the translational energy is negligible. The molecules of an ideal gas move independently of each other and the intermolecular potential energy may be neglected. For each of these cases it is possible to derive a partition function from which the properties of the state can be readily derived.

The liquid state presents quite a different problem. The intermolecular forces are sufficiently great to keep the molecules in the condensed phase, but considerable translational motion is allowed. However, the structure of the liquid may be treated as quasi-crystalline, where each atom is trapped, at least temporarily, in a small spherical cell. Within this cell the atom can move independently of its neighbors. On this basis the probability of a neighbor atom being found between the distances r and r + dr can be calculated. In this manner the radial density function can be derived, and the volume of the cell in which the center of a given atom is free to move can be determined.

Studies of the x-ray diffraction patterns obtained from liquid elements have revealed much information concerning the structure of the liquid state. They have led to a direct determination of the distribution of atoms around any given atom. This description of the structure of the liquid state may be correlated with and used to determine the characteristic properties of the liquid.

It is the purpose of this paper to apply an analytical radial density, or atomic distribution function as proposed by Wall[1] to experimentally determined distribution curves. The validity of the function is tested for the first time over a wide range of temperature for liquid mercury. The calculated curves are shown for comparison with those obtained by Fourier analysis of x-ray diffraction photographs. From this information the latent heats of fusion and vaporization are calculated and compared with observed values.

An attempt is made to correlate the free volume, within which the atom center is free to move, with the free surface energy of the liquid. The liquid is treated as a continuous medium permeated with microscopic cavities or holes of variable size. These holes, having a radius equal to that of the free volume, are not considered as a separate phase. Therefore the energy associated with the creation of such a hole should not be the total surface tension. An appropriately modified surface tension, operative at the surface of the hole, does lead to a formula adequately representing the surface tension over a wide range of temperature.

(1) Wall, C. N., Phys. Rev. 54, 1062 (1938)

REVIEW OF THE LITERATURE

The structure of liquid sodium was investigated by Wall[2] at temperatures of 100° and 400° C. The calculated distribution curve showed good agreement with the experimental curve for 100° C. It was found that the radius of the free volume was proportional to $T^{\frac{1}{2}}$ where T is the absolute temperature. On the basis of these two temperatures it was assumed that the variation was the same over the entire range. The latent heats of fusion and vaporization as calculated from the free volume were in accordance with observed values.

This investigation was repeated by Coulson and Rushbrooke [3] using the same experimental data[4] Wall used. They assumed that each atom could vibrate independently in a spherically symmetrical potential field, and derived a more general distribution curve. For comparison with this curve they recalculated the Wall distribution curve. However, their results did not differ greatly from those obtained by Wall.

The atomic distribution function for liquid potassium was investigated by Gingrich and Wall[5] again using the Wall theory. The curves for 70° C and 395° C were calculated. It was found that the free volume varied with temperature in the same manner as for sodium.

There have been many investigations made on the atomic distribution of liquid argon[6], both from a theoretical point of view and by

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(2) Ibid.
(3) Coulson, C. A. and G. S. Rushbrooke, Phys. Rev. 56, 1216 (1939)
(4) Tremble, F. H. and N. S. Gingrich, Phys. Rev. 53, 278 (1938)
(5) Gingrich, N. S. and C. N. Wall, Phys. Rev. 56, 336 (1939)
(6) Rushbrooke, G. S., Proc. Roy. Soc. Edinburgh 60, Pt. 2, No. 15, 182 (1939); Wall, C. N., Phys. Rev. 67, 285 (1945); Lund, L. H., J. Chem. Phys. 13, 317 (1945); Kerr, R. H. and L. H. Lund, J. Chem. Phys. 19, 50 (1951)

use of Wall's model in which a process of fitting is required.

In recent years a theory of the liquid state has been developed in which the liquid is assumed to be a continuum permeated with holes. Fürth[7] developed a hole model in which the holes were of molecular size and contained saturated vapor at the temperature of the liquid. A normal surface tension was attributed to the surface between the liquid and vapor phase. The holes were assumed to exhibit an irregular motion and to increase in size and frequency as the temperature increased. This theory gave only a very rough approximation to the actual behavior of the liquid.

Further study was made on the hole theory of liquids in which an attempt was made to calculate the radii, volumes, and number of holes contained in a mole of liquid[8]. The size of the holes was found to approximate two atomic diameters and the number of holes was about the same as the number of molecules. The internal pressure of several elements was calculated, but the results showed wide descrepancy. Calculations of latent heats of fusion and vaporization did not agree with the observed values. The hole theory of liquids as developed by Furth was on a classical basis. More recently a development was made by quantum methods[9]. The Schrödinger equation was used to determine the energy levels of the holes. Discrete energy levels arise as a result of the quantization of the change in radius of the holes.

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(7) Fürth, R., Proc. Camb. Phil. Soc. 37, 252 (1941)
(8) Fürth, R. and L. S. Ornstein and J. M. W. Milatz, K. Akad. Amsterdam Proc. 42.2, pp. 107-117 (1939)
(9) Auluck, F. C. and D. S. Kothari, Proc. Camb. Phil. Soc. 41, 180 (1945)

DISCUSSION

The atomic distribution function[10] of a simple liquid, $4\pi r^2 \rho(r)\, dr$, gives the average number of atoms to be found in a spherical shell of radius r and thickness dr having its center on any arbitrary atom in the liquid. The value of this function is zero for distances from the given atom less than the atomic diameter. Several atomic centers are found at a distance approximating the diameter of the atoms. For slightly greater distances, the number of atoms in each shell decreases. At a distance of about twice the atomic diameter, there occurs another maximum. Thus the distribution function shows a series of maxima and minima in the vicinity of any arbitrary central atom. As the distance approaches several atomic diameters, the density of the atoms becomes just the value given by the volume of the shell and the number of atoms per unit volume.[11]

It is possible to obtain important information concerning the liquid structure from the atomic distribution function if the potential energy of the liquid is known. A simple model of the liquid state has been proposed by Wall[12] in which the potential energy is assumed to have a minimum value for a certain set of atomic configurations. It is further assumed that the potential energy is infinite for all other configurations of the atoms or molecules. Each atom is assumed to be trapped (at least temporarily) by its immediate neighbors in a small spherical cell. Within this cell the center of the molecule is free to move without altering the potential energy of the entire assembly.

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(10) Zernike, F. and J. A. Prins, Zeits. f. Phys., 41, 187 (1927)
(11) Hildebrand, J. H., Science 80, 125 (1934)
(12) Wall, C. N., op. cit. p. 2

At the boundaries of the cell, the potential energy is assumed to be infinite. The volume of this spherical cell is defined as the free volume for the molecule.

Using this discontinuous "box-type" potential function, Wall has developed an analytical expression for the distribution function given by(13)

$$4\pi r^2 \rho_1(r) = \frac{3N_1 r}{5\sigma r_1}\left\{1 - \frac{5(r_1 - r_2)}{4\sigma^2}\left[1 - \frac{|r_1 - r|}{2\sigma} + \frac{|r_1 - r|^3}{40\sigma^3}\right]\right\} \quad \text{Eq. 1}$$

where the value of r is restricted to the interval defined by $r_1 - 2\sigma \leq r \leq r_1 + 2\sigma$ and $4\pi r^2 \rho_1(r) = 0$ for all other values of r. In the expression above, the symbols have the following meaning:

N_1 is the average number of nearest neighbors surrounding any atom in the liquid.

r_1 is the distance from the given central atom to the first maximum in the distribution curve.

$\rho_1(r)$ is the density of the atoms whose centers lie on the first coordination sphere of radius r_1.

σ is the radius of the spherical cell of free volume within which the central atom is trapped by the immediate neighbors.

In order to extend the range of the distribution curve, it is necessary to consider density contributions from coordination spheres of order higher than the first. If ρ_i is the density of atoms on the i^{th} coordination sphere and N_i is the number of atoms whose centers lie

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(13) Ibid.

on the sphere, the distribution function is given by[14]

$$4\pi r^2 \rho(r) = \sum_i 4\pi r^2 \rho_i(r) \qquad \text{Eq. 2}$$

Here $\rho_i(r)$ is defined by equations of the form given in Eq. 1 with N_1 and r_1 replaced by N_i and r_i. However, the parameters N_i and r_i are unknown functions of N_1 and r_1 and the entire curve cannot be determined analytically at this time.

The parameters of Eq. 1 can be determined by making use of the x-ray diffraction data from liquids. This function has not yet, as far as the author is aware, been applied over a wide temperature range. However, experimental distribution curves are now available for a liquid over a wide range of temperature, since the experimental distribution curves for liquid mercury have been obtained by Campbell and Hildebrand[15] for temperatures ranging from -38° C to 200° C; and these curves appear to be admirably suited, through their wide temperature range, for a thorough test of the Wall model.

For this purpose it is more convenient to use the expression $4\pi r \rho_i(r)$ rather than $4\pi r^2 \rho_i(r)$. Making this change and writing y for $4\pi r \rho(r)$, the expression in Eq. 1 becomes

$$\sigma^2 = \frac{y_1}{y_1 - y} \frac{5(r_1 - r)^2}{4} \left[1 - \frac{|r_1 - r|}{2\sigma} + \frac{|r_1 - r|^3}{40\sigma^3} \right] \qquad \text{Eq. 3}$$

where y_1 is the value of y when $r = r_1$ and it has the value $\frac{3N_1}{5\sigma r_1}$.

From the experimental data the values of y are plotted as a function of r for each temperature under consideration. The values of r_1

(14) Ibid.
(15) Campbell, J. A. and J. H. Hildebrand, J. Chem. Phys. 11, 330 (1943)

and y_1 are determined directly from these curves. The assumption is made that contributions from coordination shells of order higher than the first will be negligible in the range of $r < r_1$. Hence, the Wall distribution function may be presumed approximately valid for $r \leq r_1$.

By substituting corresponding values of y and r into Eq. 3, the value of σ may be found by a method of iteration. This is done for several values of $r < r_1$, and the average value of σ is found. The above procedure is repeated for each temperature and the results are shown in Table 1.

The approximate temperature dependence of σ is determined from the calculated average values. The best fit is obtained by using the relation

$$\sigma = 0.0097 T^{0.62} \qquad \text{Eq. 4}$$

where T is the absolute temperature and σ is obtained in angstroms. The variation of σ with temperature by Eq. 4 is shown in Fig. 1 along with values of σ from Table 1. It seems then, that σ cannot be represented as proportional to $T^{\frac{1}{2}}$ as found for the cases of Na and K.

In order to make a comparison with the experimental data, the average values of σ are used to calculate values of the function $4\pi r \rho(r)$ for each temperature. The results of this comparison are shown in Fig. 2, where the theoretical curves are shown dotted. The calculated curves compare favorably with those obtained experimentally. However, at the lower temperatures the calculated curves have a different slope. It is difficult to say whether or not this difference should be attributed to the failure of the simple model to give a true picture of the liquid.

Table 1

Calculated values of the free volume radius of liquid mercury

r in A	Sigma in A					
t°C	-38	0	50	100	150	200
2.45	0.34	0.37	0.37	0.42	0.42	0.48
2.50	0.35	0.37	0.36	0.42	0.42	0.46
2.55	0.36	0.36	0.34	0.42	0.41	0.44
2.60	0.35	0.34	0.33	0.41	0.40	0.43
2.65	0.34	0.33	0.32	0.41	0.40	0.42
2.70	0.33	0.31	0.32	0.40	0.40	0.43
2.75	0.31	0.29	0.32	0.40	0.40	0.45
2.80	0.28	0.26	0.31	0.40	0.41	0.47
2.85	0.25	0.24	0.30	0.38	0.39	0.46
2.90	0.22	0.21	0.31	0.34	0.34	0.44
2.95	0.17	0.21	0.41	0.25	0.36	0.36
Ave. σ =	0.30	0.30	0.34	0.39	0.40	0.45

VARIATION OF σ WITH TEMPERATURE

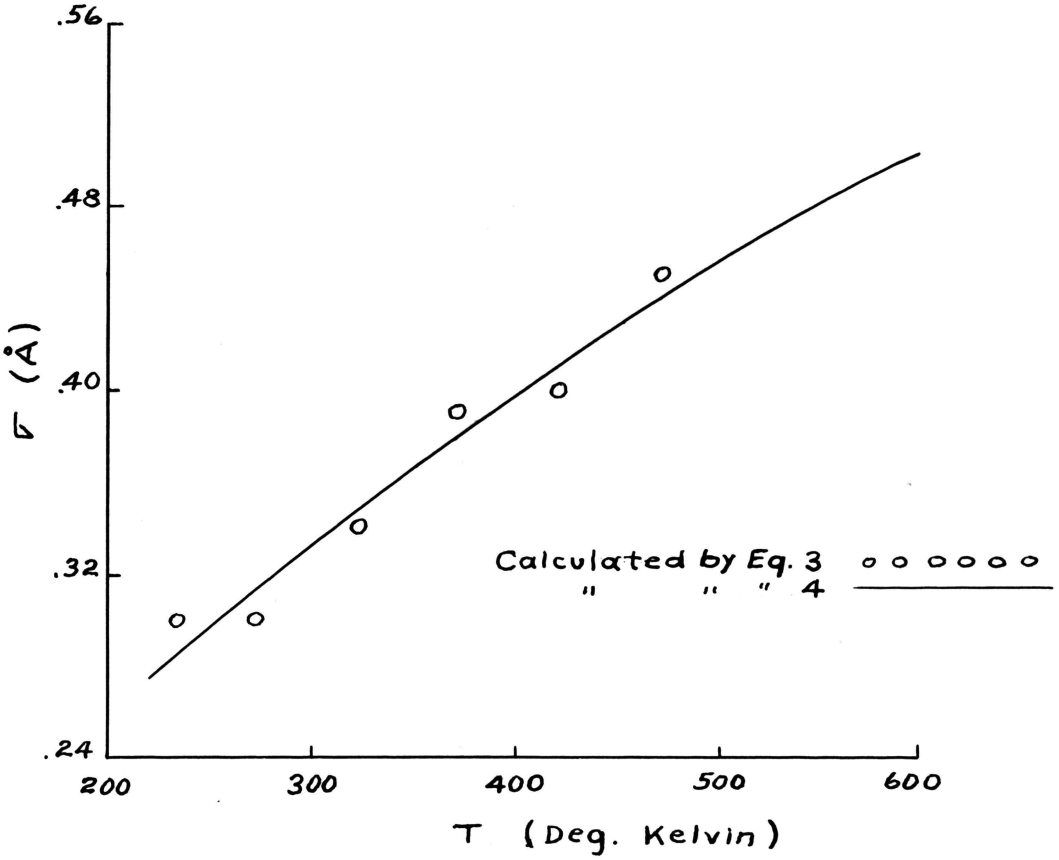

FIG. 1.

LIQUID MERCURY
DISTRIBUTION FUNCTIONS

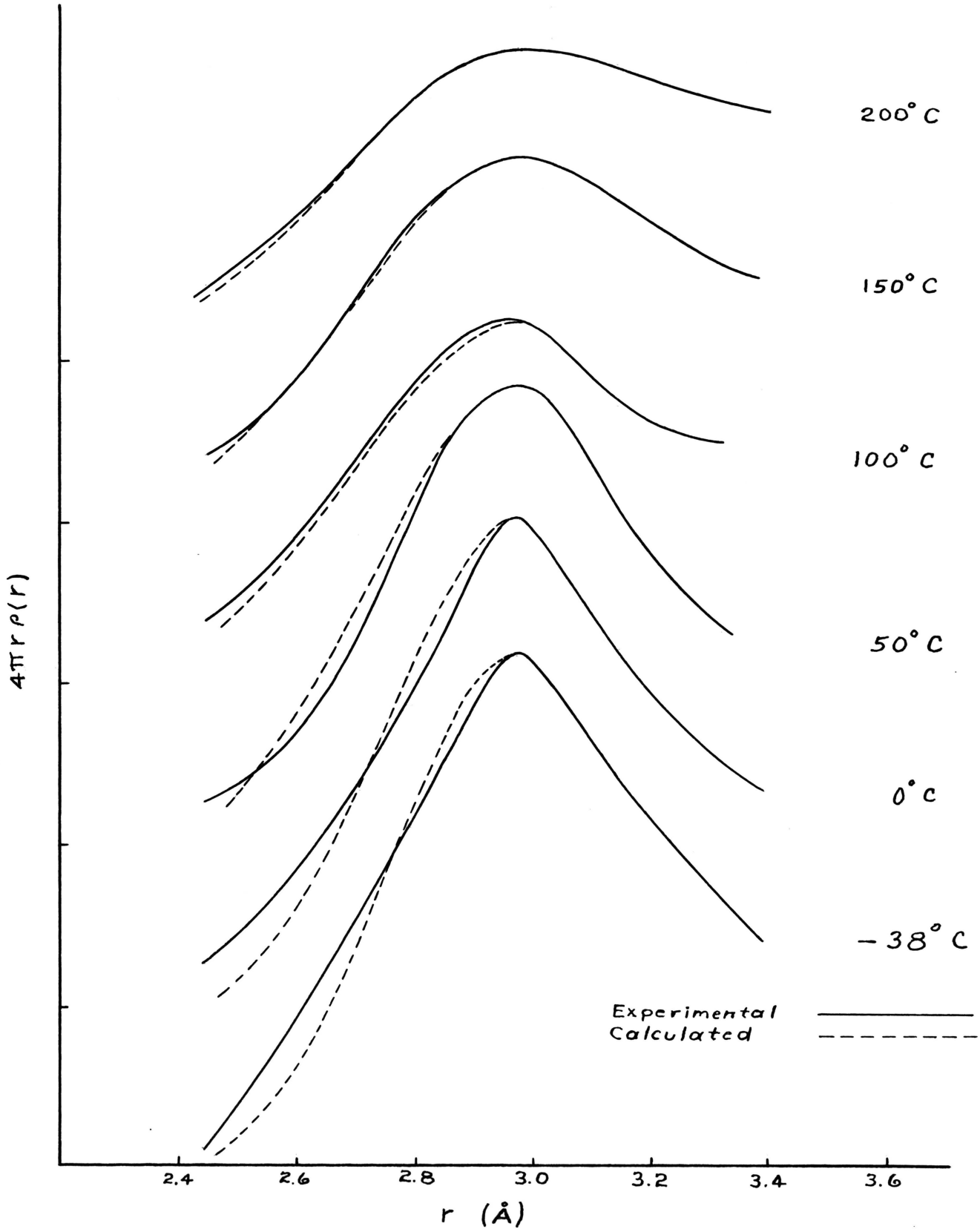

FIG. 2

One may conjecture that at higher temperatures the Wall model, which visualizes the atoms moving independently in their cells, may be more nearly correct, since at high temperatures the liquid might have a more gas-like behavior, i.e., negligible correlation in the intermolecular motion. In the case of liquid potassium[16] it was noted that the curve for 70° C fit the experimental data better than the one for 395° C.

Now the free volume per molecule can be determined from the expression for σ in Eq. 4 as $v = \frac{4}{3}\pi\sigma^3 = bT^{1.84}$ where b is determined as 3.81×10^{-30} in cgs units. This free volume can be used to calculate certain thermodynamic properties of liquid mercury. The approximate free energy of a simple liquid is given by the expression[17]

$$F = -NkT\left[\ln J(T) + \ln v\right] + \Phi \qquad \text{Eq. 5}$$

where J(T) is the ordinary partition function of the molecules, v is the free volume per molecule, and Φ is the minimum potential energy of the assembly. The other symbols have their usual meaning. For a simple monatomic liquid J(T) may be assumed to take its classical value $\left(\frac{2\pi mkT}{h^2}\right)^{\frac{3}{2}}$ where m is the mass per molecule and the other symbols have their usual significance.

The entropy of liquid mercury at the melting point, -38.87° C. can

(16) Gingrich, N. S. and C. N. Wall op. cit. p. 3
(17) Guggenheim, E. A., Proc. Roy. Soc. A135, 181 (1932)

be calculated from Eq. 5, since

$$S_{liq.} = -\left(\frac{\partial F}{\partial T}\right)_V = Nk\left[\ln\left(\frac{2\pi m k T}{h^2}\right)^{3/2} + \ln b T^{1.86} + 3.36\right] \qquad Eq.\ 6$$

This calculation gives 17 cal./mole/degree for the entropy. According to Kelley[18] the entropy of solid mercury at the same temperature is 14.5 cal./mole/degree. The entropy change at melting, 2.5 cal./mole/degree, corresponds to a latent heat of fusion equal to 2.45 kj/mole. This compares favorably with the experimental value[19] 2.33 kj/mole.

The approximate entropy of mercury vapor at the boiling point (356.9° C) can be calculated from the equation

$$S_{gas} = Nk\left[\ln\left(\frac{2\pi m k T}{h^2}\right)^{3/2} + \ln\left(\frac{V}{N}\right) + \frac{5}{2}\right] \qquad Eq.\ 7$$

where V is taken as the volume of a mole of ideal gas at the boiling point. This calculation gives S_{gas} equal to 45.5 cal./mole/degree at atmospheric pressure and 356.9° C. From Eq. 6 the entropy of the liquid at this temperature is 23.6 cal./mole/degree. The latent heat of vaporization as calculated from these values is 57.6 kj/mole. The experimental value as given in the International Critical Tables[20] is 59.3 kj/mole.

Referring back to the Wall model and to the second atom located in a spherical cell of radius σ and at a mean distance r_1 from the first, we may imagine a cavity created in the liquid of a certain volume in order that the second atom be located at its mean distance r_1 from the first. Due to the fact that the atoms are impenetrable, a spherical region of radius at least equal to $\sigma + R$ where R is the atomic radius

(18) Kelley, K. K., Bureau of Mines Bulletin 350 (1932)
(19) International Critical Tables, vol. 1 p. 102
(20) Ibid.

must be made available to the second atom. However, only a part of the region may be considered empty and capable of occupation by the center of atom 2 and that is a spherical region of radius σ. Hence, we may visualize a cavity of radius σ created in the liquid for the center of atom 2. Then we may think of the liquid as a continuous medium permeated with cavities which, at ordinary temperatures and pressures, do not have an opportunity to develop into holes of macroscopic size.

The average size of these holes and their statistical distribution over a wide range of sizes may be determined by treating the holes as if they occured in a continuous medium of constant density. For simplicity we assume the holes to be spherical in shape and to have a variable radius σ. Now the formation of a hole in the liquid is accompanied by the expenditure of a certain energy E, depending upon the size and shape of the hole. Then we may write the number of holes with radii between σ and $\sigma + d\sigma$ from classical statistics as

$$dn = N e^{-\frac{E(\sigma)}{kT}} \alpha(\sigma)\, d\sigma \qquad \text{Eq. 8}$$

where N is the total number of holes, and where it is not possible to define $\alpha(\sigma)$ uniquely, since the a priori probability to be associated with a particular value of σ cannot be ascertained.

Now at the present, there does not appear to be a way to estimate the average energy associated with the formation of such embryonic holes. If the holes were of macroscopic size, i.e. large enough to hold sufficient atoms that they could be considered to have the properties of a distinct phase, the average energy could be written as $4\pi\sigma^2\gamma$, where γ is the surface tension. Here, on such a small free

surface as we are contemplating, the full value of the surface tension would scarcely be operative. As an approximation, however, we may suppose the actual surface tension to be a certain factor, β, times the value associated with macroscopic surfaces. The validity of such an assumption can only be ascertained by the adequacy with which the physical facts can be predicted. The ensuing results indicate the assumption to be entirely tenable.

Since σ appears in the expression for energy as a quadratic factor, the theorem of equipartition of energy can be applied to obtain the average energy associated with a hole. This gives for the average energy

$$\bar{E} = \int_0^\infty E\alpha(\sigma) e^{-E/kT} d\sigma \Big/ \int_0^\infty \alpha(\sigma) e^{-E/kT} d\sigma \qquad \text{Eq. 9}$$

If we assume, for simplicity, that $\alpha(\sigma)$ is a constant, Eq. 9 can be solved for the average energy as

$$\bar{E} = \tfrac{1}{2} kT = \beta(4\pi\sigma^2\gamma) \qquad \text{Eq. 10}$$

where β is the factor by which the surface tension must be modified to apply in the case of these very small surfaces. It should be noted that this is the average energy of a hole averaged over all possible radii, and it is not an average energy associated with a translational degree of freedom.

Now the σ obtained previously as a function of T, since for a particular temperature it represents only the average radius of the free volume, may be substituted into Eq. 10 and the resulting expression for surface tension is

$$\gamma = \frac{k}{8\pi\beta(0.0097 \times 10^{-8})^2 T^{0.24}} \qquad \text{Eq. 11}$$

The constant term β can be evaluated approximately by substituting observed values of surface tension at the absolute temperature T into Eq. 11. This was done for eight observed values of surface tension[21] and the results of these calculations are shown in Table 2. It should be noted that the quantity $\frac{1}{\beta}$ has a very nearly constant value. The average was found to be 3.21 and the final expression for surface tension is

$$\gamma = \frac{1800}{T^{0.24}} \qquad \text{Eq. 12}$$

where T is in degrees Kelvin and γ is expressed as dynes per centimeter. The values of surface tension calculated from Eq. 12 are shown plotted versus temperature in Fig. 3 along with experimental values for comparison. The experimental values are predicted very well by this function. The consistent values of β seem to indicate the validity of the assumptions made. However, it should be noted that the expression for surface tension obtained here is not valid in the vicinity of the critical temperature for liquid mercury, since the notion of definite trapping of atoms in spherical cells would be highly unlikely at elevated temperatures. Nor would we expect the Wall distribution functions to agree particularly well with the experimental curves at these elevated temperatures. However, they are adequate in the range of temperatures considered here. (See Fig. 2.)

(21) Harkness, W. D. and W. W. Ewing, Am. Chem. Soc. Jour. 42, 2539 (1920)
Hogness, T. R., Am. Chem. Soc. Jour. 43, 1621 (1921)

Table 2

Values of surface tension factor $1/\beta$

	t° C	Obs. surface tension dynes/cm.	$1/\beta$
*	0	480.3	3.15
*	30	474.2	3.20
*	40	471.3	3.20
*	60	467.1	3.21
	155	441.0	3.22
	195	434.0	3.24
	200	436.0	3.27
	354	394.0	3.16
		Ave.	3.21

* Values of surface tension obtained by Harkness and Ewing, ref. (21)
Other values obtained by Hogness, ref. (21).

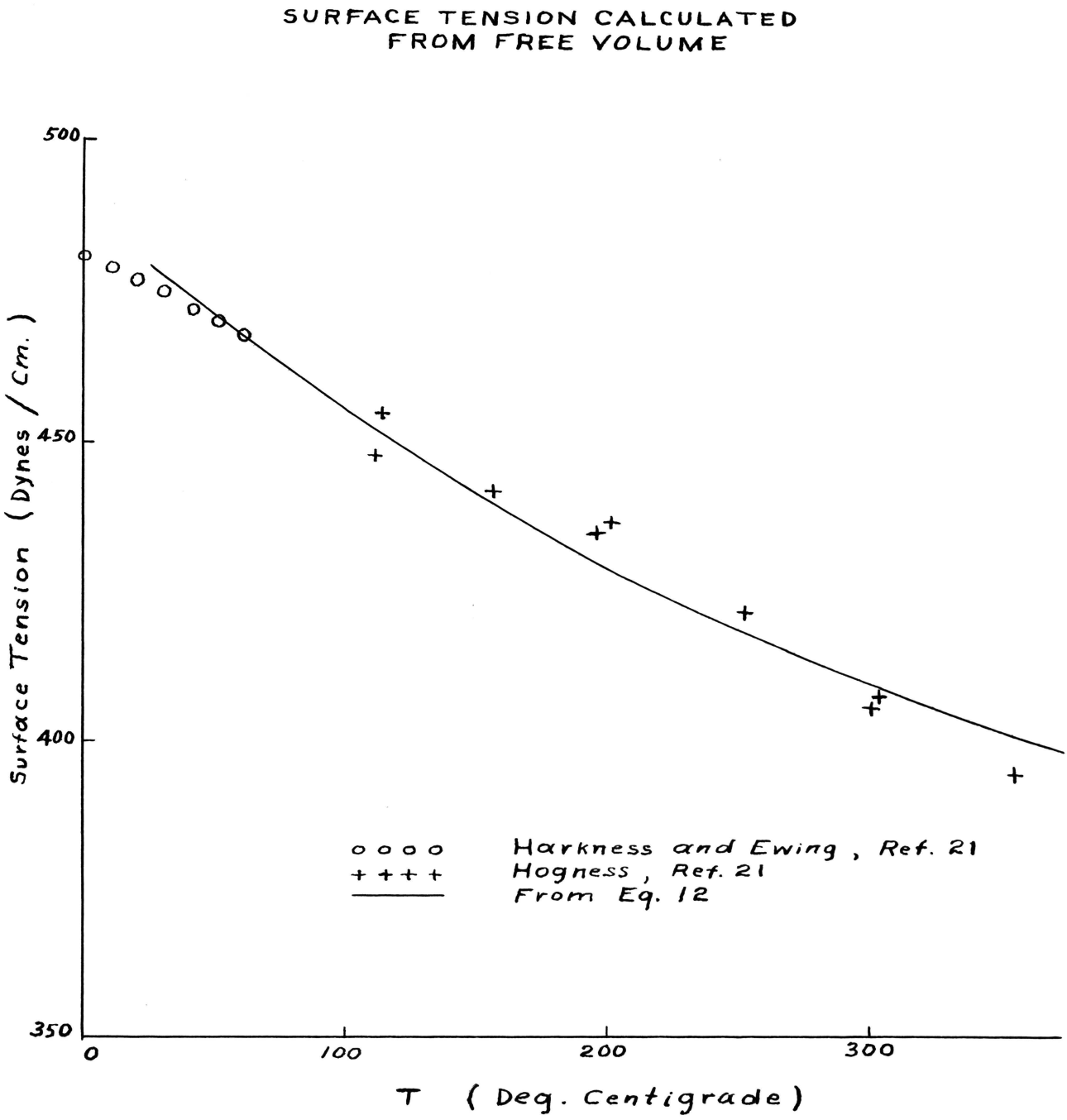
SURFACE TENSION CALCULATED
FROM FREE VOLUME
500
450
400
350
Surface Tension (Dynes / Cm.)
0
100
200
300
T (Deg. Centigrade)
o o o o Harkness and Ewing, Ref. 21
+ + + + Hogness, Ref. 21
From Eq. 12

FIG. 3.

CONCLUSIONS

The experimental distribution curves for liquid mercury have been used to test the theoretical distribution function proposed by Wall over a wide range of temperature. The calculated curves appear to fit better at the higher temperatures. This could possibly mean that the crude model used here gives a nearly correct picture of the structure at high temperatures. However, near the melting point the intermolecular forces become greater and the model, since it ignores correlation between neighboring cells at all temperatures, is not flexible enough to account for this.

The free volume for liquid mercury was found to increase with temperature as should be expected, and its functional relationship was established. However, the variation of free volume with temperature for liquid mercury does not agree with the results obtained for sodium[22] and potassium[23]. But in each case the results are based upon only two temperatures, while here a wide range of temperature was used.

The usefulness of the expression for free volume was illustrated by applying it in the calculation of the surface tension of liquid mercury at different temperatures. The close agreement of Eq. 12 with the experimental data for liquid mercury (See Fig. 3) indicates the general validity of attributing the Wall free volume to be essentially a micro-hole with a modified surface tension operative at the surface. This method can be applied to other liquids for which experimental distribution curves are available for several temperatures.

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(22) Wall, C. N., op. cit. p. 2
(23) Gingrich, N. S. and C. N. Wall, op. cit. p. 3

SUMMARY

The free volume per molecule was determined for liquid mercury by means of the Wall atomic distribution function. The temperature dependence of v_f, the radius of the free volume, was found to be given approximately by the relation $v_f = 0.00977 T^{0.62}$. The distribution curves calculated for the temperatures -38, 0, 50, 100, 150, and 200° C were plotted for comparison with the experimental curves.

Using the above relation for v_f, the entropy of liquid mercury at the melting point was found to be 17 cal./mole/deg. The heat of fusion was calculated as 586 cal./mole which is equivalent to 2.45 kj/mole. This is to be compared with the observed value of 2.33 kj/mole. The entropy of the liquid at the boiling point was determined as 23.6 cal./mole/deg. and the approximate entropy of the vapor at this temperature was found to be 45.5 cal./mole/deg. The corresponding latent heat of vaporization was 57.6 kj/mole. This is to be compared with the observed value of 59.3 kj/mole.

By treating the free volume as holes in a continuous medium, the approximate surface tension of liquid mercury was calculated. It was found that to a good approximation the surface tension was given by $\gamma = \frac{1880}{T^{0.24}}$ where T is the absolute temperature and γ is obtained in dynes per centimeter. The validity of this expression is limited to the temperature range considered here. Near the critical temperature the assumptions made in developing the Wall model and in deriving the expression for γ are not correct.

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VITA

Robert Lee Choate was born on November 30, 1918 in the town of Reevesville, Illinois. He received his elementary schooling in the public schools of the State of Illinois, and graduated from the New Columbia Community High School in May, 1939.

From March, 1942 to November, 1945 he served in the U. S. Army. In September, 1946 the writer enrolled in the Missouri School of Mines and Metallurgy and graduated in January, 1950 with the degree of Bachelor of Science in Electrical Engineering. He enrolled in the Graduate School as a candidate for the degree of Master of Science, Physics Major in January, 1950.